

WEAKLY NONLOCAL IRREVERSIBLE THERMODYNAMICS

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Dedicated to the memory prof. István Gyarmati.

ABSTRACT. Weakly nonlocal thermodynamic theories are critically revisited. A relocalized, irreversible thermodynamic theory of nonlocal phenomena is given, based on a modified form of the entropy current and new kind of internal variables, the so called *current multipliers*. The treatment is restricted to deal with nonlocality connected to dynamic thermodynamic variables. Several classical equations are derived, including Guyer-Krumhansl, Ginzburg-Landau and Cahn-Hilliard type equations.

1. INTRODUCTION

In the last decades there has been a continuous interest in developing generalized classical continuum theories to include memory and nonlocal effects. By *memory effects* one means non instantaneous interactions and actions in the continuum, that is the dynamics of the continuum is determined not only by the present state but also by the previous ones, the history of the continuum. By *nonlocal effects* one means that the interaction between representative volume elements is not local, the dynamics is determined not only by the closest neighbours, the influence of farther volume elements is not negligible. There are at least three basic methods to consider memory and nonlocal effects in the dynamic equations. One can construct *hypercontinuum theories* introducing new variables beside the space-time variables (see e.g. [1, 2]). In case of *strongly nonlocal* theories the constitutive functions are given by a space integral form directly calculating the effects of the farther neighborhood (see e.g. [3, 4]). The most popular are the so called *weakly nonlocal* theories, where higher order space derivatives are introduced into the governing equations than is usual in classical approaches. The original governing equations, that one wants to generalize, can be the balance equations of classical continuum physics or some other equations, too. The choice of the method is more or less a matter of convenience and experience, but without doubt the last method, the construction of weakly nonlocal continuum theories is the most attractive and popular among them [5].

The popularity of weakly nonlocal theories is partially based on the convenience and the familiar knowledge of differential equations. The other approaches are somehow too general to be easily understandable and manageable, and bear some basic controversies. First of all, in hypercontinua or in the strongly nonlocal approach some requirements of a physical theory (e.g. objectivity, distinction between material and structural components) are not included and exploited. Moreover, the nonlocal effects (e.g. a kind of weakening of interactions) are to be connected to the

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Second Law at least in a general heuristical sense. Seemingly there is no clear conceptual prescription in the different formulations of the Second Law that would give a firm method in hypercontinua or strongly nonlocal theories. On the other hand, weakly nonlocal theories have an approximative character. First we can consider derivatives with one order higher than in the traditional equations and we can check the effects experimentally and theoretically. More systematic considerations should give a series of weakly nonlocal equations as a kind of successive approximation.

It is interesting and instructive to review briefly the structure of the theories describing memory effects and compare them with nonlocal theories. The general memory functional method of rational thermodynamics (*strong history dependence*) (see e.g. in [6]) is the counterpart of strongly nonlocal theories. The time related counterparts of weakly nonlocal theories (*weak history dependence*) are the models that use rate dependent constitutive relations in the basic balance laws. Both are well treated and used approaches in this respect. However, the last development (more than thirty years ago) was the introduction of internal variables of different kind, and in this way one can understand that the balance structure and the Second Law can generate the rate dependence (see e.g. in [7]). This method preserves the clear physical structure of classical irreversible thermodynamics and gives a considerable flexibility in modelling physical phenomena. Let us recognize that using internal variables instead of time derivatives in the constitutive relations, the constitutive relations themselves are strictly instantaneous, they depend only on state variables of the material, therefore only on the present state. The internal variables are related to the instantaneous state of the material, they are 'local' (in time), therefore, with their help the memory effects are modelled with a local theory, we can say that they are *relocalized*. The next table summarizes the situation:

	Space	Time
Strongly nonlocal	space integrals	memory functionals
Weakly nonlocal	gradient dependent constitutive functions	rate dependent constitutive relations
Relocalized	???	internal variables

In this paper we investigate the possibility to fill the place of the question marks in the table, to relocalize weak nonlocalities by introducing new kind of internal variables in a way that is compatible with the Second Law and with traditional methods of irreversible thermodynamics. We can accomplish our task when the nonlocality is connected to the so called dynamic internal variables (dynamic degrees of freedom), those that vanish in (local) thermodynamic equilibrium.

The general strategy of weakly nonlocal approaches is to include space derivatives, gradients of field quantities in the different classical constitutive functions (therefore sometimes they are called 'gradient theories'). The justification of these generalizations is based on either checking the consequences and/or checking the requirements of basic physical principles as, for example, different forms of objectivity or/and the Second Law. Several approaches are used in mechanics e.g. different gradient theories [8, 9, 10, 11, 12, 13, 14], the virtual power considerations [15, 16] and the multifield theories [17, 18]. Instead of detailed reviewing and criticising the

different weakly nonlocal approaches our general remark is, that they usually introduce new and disputable concepts that seem to be too special to serve as foundation of a general nonlocal thermodynamic theory. Moreover, they are not constructed as a general approach to deal with nonlocal extensions of the transport equations of classical irreversible thermodynamics, they usually give methods for special fields of continuum physics (heat conduction, thermomechanics, etc..).

1.1. Requirements of the Second Law. Introducing gradient dependence in an ad-hoc way one can get equations with solutions contradicting with the Second Law. Even if we keep the equations we should exclude some initial conditions and solutions because of physical reasons (e.g. because they contradict the experiments). We should and can avoid these difficulties applying a systematic approach, a constitutive theory. To formulate a constitutive theory for nonlocal effects we strongly need a clear formulation of the Second Law and a construction method to get equations that automatically fulfill this requirement. The fundamental method of continuum physics to handle the situation is to separate the material and the structural properties of the phenomena: one should use the basic balance equations of the considered material and introduce constitutive functions that correspond to the Second Law. The question arises: what kind of Second Law, which formulation is the most fruitful? In this paper we accept that there is an extensive entropy function s , that should have a non negative production for supply free processes, in pure dissipative systems. We will see that this conservative standpoint is sufficiently general for our purposes. We do not need to weaken the well founded theoretical basis of irreversible thermodynamics and to exploit a freedom of any more general approach, e.g. to introduce a nonextensive entropy function.

In non-equilibrium thermodynamics there are three traditional methods to check the compatibility of constitutive function with the inequality of the Second Law.

- *Irreversible thermodynamics* derives an expression of the entropy production and introduces thermodynamic forces and currents with linear or non-linear relations between them [19, 20].
- *Coleman-Noll procedure* investigates more systematically the linear independence of the different possible forces and currents [21].
- *Liu procedure* looks for the form of the constitutive functions that ensures the validity of the entropy inequality with any solutions of the governing differential equations, without substituting them into the entropy balance [22].

The methods are ordered with growing exactness. The first method is constructive. One can find a more detailed treatment regarding the different formulations and exploitations of the Second Law inequality for example in [23, 24, 25]. In the following considerations one of our basic aims is to combine the heuristic power of the traditional force-current systems with a mathematically clearer treatment.

1.2. Discrete and continuum. Beyond the compatibility with the Second Law in all continuum thermodynamic theories is important and instructive to clarify the relation between discrete and continuum descriptions of physical systems. Some continuum presentations pronounce to be self standing continuum theories without any connection to thermodynamics of homogeneous, discrete systems. Homogeneous bodies are considered as special cases of continua. However, the relation is far more difficult if we consider processes in homogeneous thermodynamic systems

as it was shown in [26]. Anyway, a conceptual classification gives the basic heuristics. The relation of continuum and discrete systems is based on *continuization principles*. Classical irreversible thermodynamics is based on the *principle of local equilibrium* [19, 27], i.e. in that theory we suppose that

- physical quantities defined in ordinary (equilibrium) thermodynamics of discrete systems and the relations between them are valid in a continuum theory as well.

Particularly it means that in theories using the principle of local equilibrium the entropy function depends only on equilibrium state variables and the entropy current is classical.

In an internal variable theory we use the *principle of local state* of Kestin [28, 29], implicitly or explicitly supposing that

- physical quantities defined in non-equilibrium thermodynamics of discrete systems and the relations between them are valid in a continuum theory, too.

Particularly it means that in theories using the principle of local state the entropy function can depend on internal variables beyond the equilibrium state variables and the entropy current is classical. In this way we can get equations incorporating higher order time derivatives and we can preserve the stability structure of thermodynamics.

Let us remark that in the literature both concepts appear in a controversial treatment (including the mentioned references). For example de Groot and Mazur formulates the concept of "local equilibrium" as follows:

"It will be now assumed that, although the total system is not in equilibrium, there exists within small mass elements a state of "local" equilibrium, for which the local entropy s is the same function previously defined (for equilibrium systems) of u , v and c_k as in real equilibrium." [19]

The controversy is contained in the concept of equilibrium. A correct formulation and understanding incorporates a suitable nonequilibrium theory of discrete (homogeneous) thermodynamic bodies including internal variables started by Onsager and Machlup [30, 31] and continued by others [26, 32, 33, 34, 35, 36, 37], because in the above statement an equilibrium system is necessarily time dependent.

In nonlocal theories we are beyond the validity of local equilibrium and local state and we deal with continuum systems that do not have a discrete counterpart. In the following we introduce new kind of internal variables to characterize the nonlocality of the continuum. The nonlocal internal variables are current-like in some sense, and the extension of the state space appears not only through the modified entropy function but also through a modified entropy current. The local character of the theory is preserved because the entropy function and the entropy current function are local, do not depend on space derivatives.

The necessity to use a generalized entropy current was pointed out and emphasized several times by Müller [38, 39]. Regarding the internal variables it was Verhás who developed Müller's generalization and introduced higher order corrections with the help of transport of internal variables [40]. The importance of the entropy current describing nonlocal effects was pointed out by several previously mentioned authors [39, 41, 42].

Our approach will be called *weakly nonlocal irreversible thermodynamics* because we give conditions to transform the entropy inequality into a solvable, quadratic

form. In this way the constitutive theory can fully preserve the predictive character of irreversible thermodynamics. In this case the constitutive theory is particularly easy, one can identify force-current systems with classical, well established methods [19, 20]. The applicability of the approach is demonstrated by deriving several particular classical examples like weakly nonlocal heat conduction, generalized Ginzburg-Landau and Cahn-Hilliard type equations.

In the next section we use Liu procedure with a precise prescription of the state space, that is free from several weaknesses of the traditional treatment. In this way the background and the validity of our approach is clear and we will see among others why and in what sense Ginzburg-Landau-like and Cahn-Hilliard-like equations play a distinguished role in weakly nonlocal irreversible thermodynamics. In the third section we treat more examples and some other problems by a more heuristic approach to show the easy applicability. The last section contains a summary and discussion.

2. WEAKLY NONLOCAL IRREVERSIBLE THERMODYNAMICS

There are some basic problems in the heuristic approach of irreversible thermodynamics that should be addressed in a consistent treatment:

- One should decide what are the constitutive functions and what are the variables (derivatives) they are depending on. That is, we should fix the variables and the level of the approximation *before* the calculations, we cannot do it on the fly.
- A given, fixed state space and constitutive quantities still does not warrant that the entropy production can be written in a special quadratic form, as a sum of products of constitutive and fixed functions of the state space. One should investigate and clarify what are the conditions of that force-current structure. We will see, that in classical treatments (classical irreversible thermodynamics, classical extended irreversible thermodynamics) the form of the entropy current function is an important condition and we will investigate whether there are other choices in the considered particular systems.

In this section we treat the above questions in a systematic treatment. We accept that the entropy is an extensive quantity, therefore there is an entropy balance and we postulate that the Second Law ensures a nonnegative entropy production. We will determine what are the state variables and what are the constitutive quantities in a thermodynamic theory. E.g. the entropy and the entropy current are always constitutive, their evolution is determined by the dynamics of other physical quantities. Our task is to get restrictions that a positive entropy production and the other known restrictions (e.g. balance like dynamics) results in the form of constitutive functions, hence in the dynamics of the physical quantities. We apply the Liu theorem, given shortly in a convenient form in Appendix A. First we treat classical theories and we generalize them extending the constitutive spaces to include nonlocal effects.

We suppose that the continuum is characterized by physical quantities given by variables defined on the *basic state space* Z and therefore a local basic state of the continuum is given by $\mathbf{z} \in Z$. The basic state space is called in a slightly different context as the space of *wanted fields* [43]. It is convenient and sufficiently general to assume that the basic state space is a finite-dimensional vector space. The processes are space-time functions on the basic state space. Therefore in a nonrelativistic

treatment, for a given observer a process is $\mathbf{z}_p : \mathbb{R} \times \mathbb{R}^3 \rightarrow Z$, $(t, \mathbf{x}) \mapsto \mathbf{z}_p(t, \mathbf{x})$. We are to give the dynamic equations determining the processes, therefore we omit the treatment of the structure of the process space, because it is determined by the dynamic equations. The processes are supposed to be continuously differentiable as many times as it is necessary. Moreover, we are fully aware of the fact that the structure of space-time is more refined also in the nonrelativistic case [44, 45], but for the sake of simplicity and easier understanding we restrict ourselves to the level of treatment usual in the literature of the field. One should be careful in applying our following considerations to mechanical theories, because mechanics is strongly connected to the structure of space-time [46]. To emphasize formally the necessary care, the balances are written in a local frame, relatively to an inertial observer. Without any further considerations they are valid in a continuum at rest respectively to that observer.

We postulate that the entropy is an extensive quantity with non negative production. This can be formulated in the form of a local balance equation

$$(1) \quad \partial_t s + \nabla \cdot \mathbf{j}_s = \sigma_s \geq 0.$$

∂_t denotes the partial derivative with respect to time. The entropy density s and the entropy current \mathbf{j}_s are considered as constitutive quantities, hence the nonnegative entropy production σ_s have to be a consequence of pure material properties. We assume that the above inequality expresses a pure material property and is fulfilled independently on the initial and boundary conditions of the governing equations (see [25]). We look for constitutive relations with the help of Liu's theorem to ensure that nonnegativity .

In addition of the entropy and the entropy current there can be other constitutive quantities in the theory. The domain of the constitutive functions is called *constitutive state space*. The particular theories are determined basically by the choice of the basic state space and the constitutive state space.

2.1. Classical irreversible thermodynamics. In classical irreversible thermodynamics the *basic state space* Z_{CIT} is spanned by the specific extensive variables. Therefore a local basic state of the continuum is given as an element of this space $\mathbf{a} = (a_1, a_2, \dots, a_n) \in Z_{CIT}$, where n is the number of considered extensive quantities. The balance expressing the extensivity of the variables is written as follows

$$(2) \quad \partial_t \mathbf{a} + \nabla \cdot \mathbf{j}_a = \sigma_a.$$

Here the current density of the extensives $\mathbf{j}_a = (\mathbf{j}_1, \mathbf{j}_2, \dots, \mathbf{j}_n)$ is a direct product vector of the conductive current densities respectively and is a constitutive quantity. Every current has a tensorial order one degree higher than the corresponding specific extensive quantity. The dot between the nabla and the current denotes a contraction for that extra tensorial order. The constitutive space of classical irreversible thermodynamics is spanned by the extensives and their gradients $C_{CIT} = Z_{CIT} \times \text{Lin}(Z_{CIT}, \mathbb{R}^3)$. A *constitutive state* of the material is an element of this space $(\mathbf{a}, \nabla \mathbf{a}) \in C_{CIT}$. The so called *process direction space* is spanned by the next space and time derivatives of the basic state: $(\partial_t \mathbf{a}, \partial_t \nabla \mathbf{a}, \nabla^2 \mathbf{a})$, where ∇^2 denotes the second derivative with respect to the space variables. The constitutive quantities that depend on the space-time through the constitutive space are the entropy, the entropy current and the current density of the extensives $(s, \mathbf{j}_s, \mathbf{j}_a)$. The

subset of the constitutive space where they are defined is the *constitutive domain* $D \subset C_{CIT}$ of the classical irreversible thermodynamic material.

After these preliminary nominations we are looking for restrictions of the constitutive functions posed by the balance form dynamic equation and the nonnegativity of the entropy' production. Considering the previously given constitutive dependencies we can write (2) and (1) as

$$(3) \quad \partial_t \mathbf{a} + \partial_1 \mathbf{j}_a \cdot \nabla \mathbf{a} + \partial_2 \mathbf{j}_a : \nabla^2 \mathbf{a} - \sigma_a = \mathbf{0},$$

$$(4) \quad \partial_1 s \partial_t \mathbf{a} + \partial_2 s \partial_t \nabla \mathbf{a} + \partial_1 \mathbf{j}_s \cdot \nabla \mathbf{a} + \partial_2 \mathbf{j}_s : \nabla^2 \mathbf{a} \geq 0.$$

Here ∂_1 denotes the partial derivative of the constitutive quantities with respect to \mathbf{a} , by the first member of the constitutive space. ∂_2 is the derivative with respect to the second member, by $\nabla \mathbf{a}$. Now we are looking for constitutive functions that will ensure the validity of the entropy inequality for all possible process directions enabled by the balances.

Applying Liu's theorem to the equations above (see Appendix A) the process direction space spanned by $(\partial_t \mathbf{a}, \partial_t \nabla \mathbf{a}, \nabla^2 \mathbf{a})$ corresponds to \mathbb{V} . The space into which the balances are mapping, that is essentially the basic state space Z_{CIT} divided by the one dimensional vector space of vectorized time [44], corresponds to \mathbb{V}' . The application of the theorem results in that there exist $\Gamma \in Z_{CIT}^*$ and the related Liu-equations are

$$(5) \quad \partial_1 s - \Gamma = \mathbf{0},$$

$$(6) \quad \partial_2 s = \mathbf{0},$$

$$(7) \quad (\partial_2 \mathbf{j}_s - \Gamma \partial_2 \mathbf{j}_a)^s = \mathbf{0}.$$

The superscript s denotes the symmetric part of the corresponding second order tensor. Moreover, the dissipation inequality will be

$$\partial_1 \mathbf{j}_s \cdot \nabla \mathbf{a} - \Gamma \partial_1 \mathbf{j}_a \cdot \nabla \mathbf{a} + \Gamma \sigma_a \geq 0.$$

From the first two Liu-equations one can see immediately that the entropy is a function of the basic state \mathbf{a} and the Lagrange multiplier can be identified with the intensive variable defined by $Ds = \partial_1 s = \Gamma$. A solution of (7) results in that

$$(8) \quad \mathbf{j}_s(\mathbf{a}, \nabla \mathbf{a}) = \Gamma(\mathbf{a}) \mathbf{j}_a(\mathbf{a}, \nabla \mathbf{a}) + \mathbf{j}_0(\mathbf{a}),$$

where \mathbf{j}_0 is an arbitrary function. From these consequences of the Liu equations one can get the dissipation inequality as

$$(9) \quad \mathbf{j}_a \cdot \nabla \Gamma + \nabla \cdot \mathbf{j}_0 \geq 0.$$

If $\sigma_a = \mathbf{0}$ and $\mathbf{j}_0 = \mathbf{0}$ then the entropy current and the entropy production reduces to the well known classical form of irreversible thermodynamics

$$\mathbf{j}_s = \Gamma \mathbf{j}_a$$

and

$$\mathbf{j}_a(\mathbf{a}, \nabla \mathbf{a}) \cdot \nabla \Gamma \geq 0.$$

The general solution of the above inequality gives the classical Onsagerian conductivity relations (see Appendix C), where one can identify the current densities of extensives as thermodynamic currents and the gradients of the intensives as thermodynamic forces of classical irreversible thermodynamics.

$$\mathbf{j}_a = \mathbf{L} \nabla \Gamma$$

with positive semidefinite conductivity matrix \mathbf{L} . In general the conductivity matrix can depend on the constitutive state and is called as quasilinear. In case of constant conductivity one can say that the theory is strictly linear [47].

It is interesting to observe that the validity of classical force-current relations is connected to the condition that in the entropy inequality $\sigma_a \Gamma$ should be nonnegative separately to the other parts of the inequality. That condition is not true in general and is used to separate dissipative and nondissipative parts of the source term σ_a (For example in a thermomechanics of point-masses one can distinct between damping, conservative and exiting momentum sources, force fields.) To avoid the details of that problem we demanded that $\sigma_a = \mathbf{0}$. The systems corresponding to the above special case are sometimes called as *pure dissipative* and play a certain role in connection to variational principles of irreversible thermodynamics [48, 49].

Furthermore let us observe that the general entropy current and the source differs from the classical expressions in the term \mathbf{j}_0 . Several authors suggest an additive supplement to the classical entropy current (e.g. the celebrated \mathbf{K} vector of Müller [38]). In the following in connection to more general theories we will see that such term can have a physical significance which also cannot be excluded here (e.g. thermodynamic theories of chemical reactions).

We have seen that in our calculations the classical form of the entropy production (the first term in the inequality above) *and* the classical form of the entropy current were consequences of the Second Law, without referring to any 'local equilibrium' arguments. Formulating the possible functional dependencies of the corresponding quantities and the usual (sometimes) implicit postulates (balance structure, existence of entropy and the entropy inequality), we encountered a clear distinction between postulates and consequences and the limits of validity of the local equilibrium approximation. For example one can see, that the second half of the principle of local equilibrium is completely unnecessary, the validity of the equilibrium state functions and the classical form of the entropy current are both consequences of structural requirements.

The choice of the constitutive space looks like a crucial point in the above derivation. Remarkable, that an extension of the constitutive space with higher order space derivatives does not alter the previous scheme. A short calculation shows that in the final Onsagerian conductivity equations the conductivity coefficients can be functions of the full constitutive space, that is they can depend on all the higher order space derivatives, but without further assumptions the form of the entropy production does not change. What can make a difference is the extension of the basic state space as we will see in the next subsection.

2.2. Classical extended irreversible thermodynamics. Extended thermodynamics is a nonequilibrium thermodynamic theory, where the currents of traditional extensive quantities are introduced as internal variables. The theory is sometimes called *extended irreversible thermodynamics* [50, 51], *extended rational thermodynamics* [39] or *wave approach to thermodynamics* [7] depending on the details of the approach and the school of thermodynamicians. These details are not too important in our treatment. One of the advantages of using currents as independent variables is that in this case the final equations can be compared directly with results of the kinetic theory, with the moment series expansion of the Boltzmann equation. This comparison gives a remarkable insight into the material characteristics of gases and (partially) of fluids.

In the following we comply the procedure of the previous subsection in a somewhat shortened form. In our treatment the difference is that the basic state space of classical irreversible thermodynamics is supplemented by the space of current density of the extensives, it is spanned by the variables $(\mathbf{a}, \mathbf{j}_a) \in Z_{ET}$. Therefore, an appropriate constitutive space will be $C_{ET} = Z \times \text{Lin}(Z, \mathbb{R}^3) \times \text{Lin}(Z, \mathbb{R}^3)$ and a constitutive state of the extended irreversible material is $(\mathbf{a}, \mathbf{j}_a, \nabla \mathbf{a}) \in C_{ET}$. In this way we supposed that there should be an evolution equation for the current and we are to find a possible form of this equation, according to thermodynamic requirements. The conditions are the balances of extensives and the nonnegative entropy production. Therefore the balance (2) and the inequality (1) are supplemented by the next general form of the searched evolution equation

$$(10) \quad \partial_t \mathbf{j}_a + \mathcal{G} = \mathbf{0}.$$

The constitutive quantities defined on the constitutive space are s, \mathbf{j}_s and \mathcal{G} . Considering the constitutive dependencies, the balances and the entropy inequality appear in the form

$$\begin{aligned} \partial_t \mathbf{a} + \nabla \cdot \mathbf{j}_a &= \sigma_a, \\ \partial_t \mathbf{j}_a + \mathcal{G} &= \mathbf{0}, \\ \partial_1 s \partial_t \mathbf{a} + \partial_2 s \cdot \partial_t \mathbf{j}_a + \partial_3 s \cdot \partial_t \nabla \mathbf{a} + \partial_1 \mathbf{j}_s \cdot \nabla \mathbf{a} + \partial_2 \mathbf{j}_s : \nabla \mathbf{j}_a + \partial_3 \mathbf{j}_s : \nabla^2 \mathbf{a} &\geq 0. \end{aligned}$$

Here, according to our concise notation, the different dots are denoting the contractions of the spacelike tensorial orders (not involved in Z_{ET}). Therefore, because of Liu's theorem there exist Lagrange multipliers Γ_e and Γ_n of the first two equations so that Liu equations are

$$\begin{aligned} \partial_1 s - \Gamma_e &= \mathbf{0}, \\ \partial_2 s - \Gamma_n &= \mathbf{0}, \\ \partial_3 s &= \mathbf{0}, \\ \partial_2 \mathbf{j}_s - \Gamma_e \mathbf{I} &= \mathbf{0}, \\ (\partial_3 \mathbf{j}_s)^s &= \mathbf{0}. \end{aligned}$$

The dissipation inequality in our case is

$$(11) \quad \partial_1 \mathbf{j}_s \cdot \nabla \mathbf{a} - \Gamma_n \mathcal{G} - \Gamma_e \sigma_a \geq 0.$$

A solution of Liu equations gives that s and \mathbf{j}_s do not depend on the gradient of \mathbf{a} and the partial derivatives of the entropy give the Lagrange multipliers, $Ds(\mathbf{a}, \mathbf{j}_a) = (\partial_1 s, \partial_2 s) = (\Gamma_e, \Gamma_n)$. Thus the Liu equations give a condition for the entropy current as

$$(12) \quad \partial_2 \mathbf{j}_s(\mathbf{a}, \mathbf{j}_a) = \partial_1 s(\mathbf{a}, \mathbf{j}_a) \mathbf{I},$$

where \mathbf{I} is the unit tensor of the basic state space. The postulate of pure dissipativity ($\sigma_a = \mathbf{0}$) also simplify the dissipation inequality and we get that

$$(13) \quad \partial_1 \mathbf{j}_s \cdot \nabla \mathbf{a} - \partial_2 s \cdot \mathcal{G} \geq 0.$$

However, we cannot give a general solution of (12) and (13) without any further ado. Fortunately we can go farther, because according to theories of extended thermodynamics one can exploit some physical requirements writing our non-equilibrium entropy function in the following form

$$(14) \quad s(\mathbf{a}, \mathbf{j}_a) = s_0(\mathbf{a}) - \frac{1}{2} \mathbf{j}_a \cdot \mathbf{m}(\mathbf{a}, \mathbf{j}_a) \cdot \mathbf{j}_a,$$

where the matrix \mathbf{m} is called the *matrix of the thermodynamic inductivities*, $s_0(\mathbf{a}) = s(\mathbf{a}, \mathbf{0})$ is the *equilibrium entropy*, that depends only on the extensives. It is worth remarking that this form, with state dependent thermodynamic inductivities is not an approximation if we accept the following requirements:

- The current variables \mathbf{j}_a are nonequilibrium, dynamic variables (dynamic degrees of freedom) that identically vanish in (local) thermodynamic equilibrium. There are extensives with such currents (heat and diffusion currents) but there are also evident counterexamples (pressure). The treatment of nonlocality connected to non dynamic variables is beyond the scope of this paper.
- The entropy is a concave function. According to that requirement it is concave also in the nonequilibrium part of the state space therefore the symmetric part of \mathbf{m} is assumed to be positive definite.
- The derivative of the entropy function characterizes the equilibrium.

With this premises the mean value theorem results in (14) as the most general form of the entropy (see Appendix B). In case of general internal variables where the physical meaning is fixed only after the thermodynamic restrictions are applied, the Morse lemma can give a more restricted form of the entropy function, with constant thermodynamic inductivities [7, 52]. However, in extended thermodynamics the current densities cannot be transformed arbitrarily, therefore a constant inductivity matrix would be an approximation.

The form (14) of non-equilibrium entropy was introduced first by Machlup and Onsager for discrete systems using the time derivatives of the original α quantities as dynamic variables [31]. It was Gyarmati who suggested the above form for field theories using the current densities as dynamic variables [47]. This kind of entropy function was originally a characteristic constituent of the wave approach of thermodynamics, the other theories realized the principle of local state through a generalization of the Gibbs relation.

The previously introduced non-equilibrium intensives are the following

$$(15) \quad \Gamma_e(\mathbf{a}, \mathbf{j}_a) := \frac{\partial s}{\partial \mathbf{a}} = \frac{\partial s_0}{\partial \mathbf{a}} - \frac{1}{2} \mathbf{j}_a \cdot \frac{\partial \mathbf{m}}{\partial \mathbf{a}} \cdot \mathbf{j}_a,$$

$$(16) \quad \Gamma_n(\mathbf{a}, \mathbf{j}_a) := \frac{\partial s}{\partial \mathbf{j}_a} = - \left(\mathbf{m}^s + \frac{1}{2} \mathbf{j}_a \cdot \frac{\partial \mathbf{m}}{\partial \mathbf{j}_a} \right) \cdot \mathbf{j}_a =: -\hat{\mathbf{m}} \cdot \mathbf{j}_a,$$

where the superscript s denotes the symmetric part of the corresponding quantity and $\hat{\mathbf{m}}$ is a nonequilibrium inductivity. $\hat{\mathbf{m}}$ is symmetric if \mathbf{m} is constant.

The form (14) is still too general to solve (12) and (13) without further assumptions, therefore we restrict ourselves to the usual approximation, when the matrix of thermodynamic inductivities is constant:

$$s(\mathbf{a}, \mathbf{j}_a) = s_0(\mathbf{a}) - \frac{1}{2} \mathbf{j}_a \cdot \mathbf{m} \cdot \mathbf{j}_a.$$

In this case the solution of (12) is similar to that we have got in the previous subsection

$$\mathbf{j}_s(\mathbf{a}, \mathbf{j}_a) = \partial_1 s \mathbf{j}_a + \mathbf{j}_0(\mathbf{a}).$$

The dissipation inequality becomes

$$(\mathcal{G} \cdot \mathbf{m}^s + \nabla \partial_1 s) \cdot \mathbf{j}_a \geq 0.$$

We have got a product of fixed functions of the basic state and constitutive quantities to be determined. The general solution of this kind of inequalities is treated shortly in Appendix C. Here we have got constitutive expressions for \mathcal{G} as Onsagerian conductivity relations. Therefore, the form of the evolution equation of the current densities is determined by

$$\mathbf{j}_a = \mathbf{L}(\mathcal{G} \cdot \mathbf{m}^s + \nabla \partial_1 s).$$

Here \mathbf{L} is a positive semidefinite conductivity matrix, depending on the constitutive state. Substituting into (10) we get

$$\partial_t \mathbf{j}_a + \mathbf{m}^{s-1}(\mathbf{L}^{-1} \mathbf{j}_a - \nabla \partial_1 s) = \mathbf{0}.$$

2.3. Weakly nonlocal extended irreversible thermodynamics. Our next example is the first weakly nonlocal extension of extended irreversible thermodynamics. The basic state space is the same as in the last subsection but we will consider an extension of the constitutive space. Here we need the first and second derivatives of the current densities, too. Hence the constitutive space of nonlocal extended thermodynamics is $C_{NET} = Z \times \text{Lin}(Z, \mathbb{R}^3) \times \text{Lin}(Z, \mathbb{R}^3) \times \text{Lin}(Z, \mathbb{R}^6) \times \text{Lin}(Z, \mathbb{R}^9)$ and the constitutive functions will depend on the quantities $(\mathbf{a}, \mathbf{j}_a, \nabla \mathbf{a}, \nabla \mathbf{j}_a, \nabla^2 \mathbf{j}_a) \in C_{NET}$. The balance of the extensives (1), the evolution equation of the currents (10) and the entropy inequality are to be solved.

In classical irreversible thermodynamics and extended irreversible thermodynamics the choice of the state space made possible to prove the local equilibrium and local state hypothesis: the entropy function did not depend on the nonlocal part of the constitutive space, on the gradient of the extensive quantities. In our case, the above extension of the state space does not enable such kind of proof, therefore we will postulate it. This is an assumption, we will call that the *hypothesis of relocatability*. However, instead of directly prescribing that the entropy does not depend on the gradients we make a seemingly weaker hypothesis requiring that the gradient of the current evolution equation (10) does not appear among the constraints. Furthermore, we treat pure dissipative systems with dynamic current densities.

The constitutive quantities defined on the constitutive space are the same as previously s, \mathbf{j}_s and \mathcal{G} . Accordingly, the balances and the entropy inequality appear in the form

$$\begin{aligned} \partial_t \mathbf{a} + \nabla \cdot \mathbf{j}_a &= \mathbf{0}, \\ \partial_t \mathbf{j}_a + \mathcal{G} &= \mathbf{0}, \\ \partial_1 s \partial_t \mathbf{a} + \partial_2 s \cdot \partial_t \mathbf{j}_a + \partial_3 s \cdot \partial_t \nabla \mathbf{a} + \partial_4 s : \partial_t \nabla \mathbf{j}_a + \partial_5 s \cdot : \partial_t \nabla^2 \mathbf{j}_a + \\ \partial_1 \mathbf{j}_s \cdot \nabla \mathbf{a} + \partial_2 \mathbf{j}_s : \nabla \mathbf{j}_a + \partial_3 \mathbf{j}_s : \nabla^2 \mathbf{a} + \partial_4 \mathbf{j}_s \cdot : \nabla^2 \mathbf{j}_a + \partial_5 \mathbf{j}_s :: \nabla^3 \mathbf{j}_a &\geq 0. \end{aligned}$$

Introducing the Lagrange multipliers Γ_e and Γ_n for the first two equalities, the Liu equations are

$$\begin{aligned} \partial_1 s - \Gamma_e &= 0, \\ \partial_2 s - \Gamma_n &= 0, \\ \partial_i s &= 0, \quad i = 3, 4 \\ (\partial_5 s)^s &= 0, \\ (\partial_i \mathbf{j}_s)^s &= 0, \quad i = 3, 5. \end{aligned}$$

Here the superscript s of $\partial_5 \mathbf{j}_s$ can be expressed best with indexes that $(\partial_5)_{ijk}(\mathbf{j}_s)_l$ is symmetric in ijk . Because of the larger constitutive space the dissipation inequality is considerably longer

$$(17) \quad \partial_1 \mathbf{j}_s \cdot \nabla \mathbf{a} + (\partial_2 \mathbf{j}_s - \Gamma_e I) : \nabla \mathbf{j}_a + \partial_4 \mathbf{j}_s : \nabla^2 \mathbf{j}_a - \Gamma_n \mathcal{G} \geq 0.$$

A solution of the Liu equations gives that the entropy is a function of \mathbf{a} and \mathbf{j}_a solely and does not depend on the gradients at all (that is $s(\mathbf{a}, \mathbf{j}_a)$) and \mathbf{j}_s does not depend on the gradient of \mathbf{a} and the second gradient of \mathbf{j}_a (that is $\mathbf{j}_s(\mathbf{a}, \mathbf{j}_a, \nabla \mathbf{j}_a)$). The partial derivatives of the entropy give the Lagrange multipliers respectively. The dissipation inequality reduces to

$$\nabla \cdot \mathbf{j}_s - \Gamma_e \nabla \mathbf{j}_a - \Gamma_n \mathcal{G} \geq 0.$$

We can see that the solution of the Liu equations does not give a soluble form of the dissipation inequality. However, it is easy to transform it to the traditional force-current form after some further, not too restrictive premises. Let us turn our attention to the entropy current. A natural physical assumption is that entropy, being connected to other extensives, does not flow if the extensive quantities do not. Here we should apply the same assumption as in case of classical extended thermodynamics suppose the the current of the extensives is a *dynamic variable* and vanishes in local thermodynamic equilibrium. Therefore we can assume that the entropy current is zero if the currents of the extensive quantities are identically zero $\mathbf{j}_s(\mathbf{a}, \mathbf{0}, \mathbf{0}) = \mathbf{0}$. Considering this condition and using the mean value theorem, one can get the next functional form

$$(18) \quad \mathbf{j}_s(\mathbf{a}, \mathbf{j}_a, \nabla \mathbf{j}_a) = \mathbf{B}(\mathbf{a}, \mathbf{j}_a, \nabla \mathbf{j}_a) \cdot \mathbf{j}_a,$$

where the *current intensity factor* \mathbf{B} , can be supposed to be continuously differentiable in a neighborhood of the local equilibrium state. We should remark again that this form itself is not an approximation, it is general with the above conditions. One can observe that the tensorial order of the current intensity factor \mathbf{B} is one order higher than the corresponding current. As we mentioned above, the classical form of the entropy current was investigated and generalized by several authors. The above kind of multiplicative generalization is due to N  ri [53].

Now the entropy production can be written as

$$(19) \quad (\mathbf{B} - \Gamma_e I) : \nabla \mathbf{j}_a + (\nabla \cdot \mathbf{B} + \mathcal{G} \hat{\mathbf{m}}) \cdot \mathbf{j}_a \geq 0.$$

where $\hat{\mathbf{m}}$ is the inductivity function defined in (16). The general solution of the inequality is

$$(20) \quad \begin{aligned} \nabla \cdot \mathbf{B} - \partial_t \mathbf{j}_a \cdot \hat{\mathbf{m}} &= \mathbf{L}_{11} \mathbf{j}_a + \mathbf{L}_{12} \nabla \mathbf{j}_a, \\ \mathbf{B} - \Gamma_e I &= \mathbf{L}_{21} \mathbf{j}_a + \mathbf{L}_{22} \nabla \mathbf{j}_a. \end{aligned}$$

We can eliminate the current intensity factor and get

$$(21) \quad \hat{\mathbf{m}}^* \cdot \partial_t \mathbf{j}_a - \nabla \cdot (\mathbf{L}_{21} \mathbf{j}_a + \mathbf{L}_{22} \nabla \mathbf{j}_a + \Gamma_e I) = -(\mathbf{L}_{11} \mathbf{j}_a + \mathbf{L}_{12} \nabla \mathbf{j}_a).$$

The star denotes the transpose. One can see, that contrary to the common postulate [39] in extended thermodynamics, this evolution equation does not have a balance form in general. It could be transformed formally into a balance form with special source in case of constant inductivities.

Let us remark that a smaller extension of the constitutive space with only the gradient of the current densities would not have been enough to get a gradient dependent entropy current.

2.4. Weakly nonlocal internal variables. In the previous subsections the physical meaning of the internal variables was fixed, they were the current densities of the thermodynamic extensives. However, it is a restriction, the usage and applicability of internal variables is far more broader [54, 55]. We can meet them under different names as *order parameters*, *dynamic degrees of freedom*, *dynamic variables* in different physical theories with a slightly different meaning. They are applied characterizing materials with complex microstructure, properties of gases near critical states, etc. The extent and method of applying thermodynamic restrictions in internal variable theories can be very different in different approaches. In thermodynamic theories the application of internal variables is connected to the principle of local state and therefore to discrete systems [56]. Their dynamics is restricted or generated constitutively by the Second Law and results in ordinary differential equations. In discrete systems, i.e. in homogeneous bodies, the formalism is well known and frequently applied, however sometimes the dynamic aspects are hidden (a good summary of the dynamics of the simplest discrete thermodynamic system without internal variables is given from several point of view in [57, 58, 26] and with internal variables in [33, 35]). On the other hand, as we mentioned in the introduction, these internal variable theories can be considered as a "relocalization" of some memory, inertial effects of the equilibrium variables. The homogeneous dynamics (ordinary differential equations) in continuous systems has some important disadvantages: the local state seems to be too tight from several points of view. First of all some physical phenomena cannot be modelled with them (e.g. thickness of shear bands, structure of surfaces, ...). Moreover, the numerical solution of coupled ordinary and partial differential equations depends on the discretization, on the mesh.

The situation is entirely different in case of weakly nonlocal equations. Here one can distinguish between essentially two independent approaches. One of them is based on the fundamental *balance equations* introducing gradient dependent terms directly into the constitutive functions, which are generated using the local equilibrium or the local state hypothesis (see e.g. [59] and numerous examples in rheology and mechanics in general [60, 13, 14]).

The other approach is even more intuitive. There one assumes that the local entropy function depends on the gradients of the corresponding physical quantities (usually on a rather simple way) and a kind of 'nonlocal intensives' are generated by the functional derivatives of the entropy function. This method is used to get the Ginzburg-Landau equation for a so called 'non conservative' physical quantity (internal variable), and the Cahn-Hilliard equation for a 'conservative', extensive one (model A and B with the terminology of Hohenberg and Halperin [61]). The general equation for the nonequilibrium reversible-irreversible coupling (GENERIC) is based on a generalization of this kind of approach [62, 63].

The relation of the two approaches is not clear at all. With the traditional approach (based on local equilibrium and state) one cannot get the most important nonlocal theories of contemporary physics (e.g. Ginzburg-Landau) on the other hand the variational derivations are too intuitive and they have nothing to do with the balances of the fundamental physical quantities. Criticizing the mentioned derivations of Ginzburg-Landau and Cahn-Hilliard equations, Gurtin emphasizes the importance of the separation of balances from the constitutive properties:

"My view is that while derivations of the form ... are useful and important, they should not be regarded as basic, rather as precursors of more complete theories. While variational derivations often point the way toward a correct statement of basic laws, to me such derivations obscure the fundamental nature of balance laws in any general framework that includes dissipation." [64]

Gurtin himself uses a method exploiting his "microforce balance principle", which is a new principle again, and we can apply Gurtin's further critical remarks used against the variational approach also to his propositions:

- The derivations limit the manner how other terms (e.g. rate terms) enter the equations.
- The derivations are specific, it is not clear how they can be generalized for general thermodynamic processes including not only mechanics, diffusion or the dynamics of the order parameters near critical states.

In this section we will see how far can we go with relocalized theories, with the methods of irreversible thermodynamics. We introduce nonlocal internal variables connected to the entropy current. First we will see the weakly nonlocal irreversible thermodynamic theory of pure dynamic internal variables without further constraints, after that we investigate the case of a nonlocal extension of a thermodynamic theory containing also extensive variables.

2.4.1. Weakly nonlocal dynamic internal variable - Ginzburg-Landau equation. Our task is to find weakly nonlocal evolution equation of an internal variable that corresponds to the requirement of nonnegative entropy production. The basic state space, Z_I , is spanned by the vector $\boldsymbol{\xi}$, denoting an array of internal variables with different tensorial character. We are to find an evolution equation of the internal variable in the form:

$$(22) \quad \partial_t \boldsymbol{\xi} + \mathcal{F} = \mathbf{0}.$$

The constitutive space is spanned by $\boldsymbol{\xi}$ and its first and second gradients: $(\boldsymbol{\xi}, \nabla \boldsymbol{\xi}, \nabla^2 \boldsymbol{\xi})$. Therefore $C_I = Z_\xi \times \text{Lin}(Z_\xi, \mathbb{R}^3) \times \text{Lin}(Z_\xi, \mathbb{R}^6)$ is the constitutive space of the nonlocal dynamics of an arbitrary internal variable. The constitutive quantities are the entropy, the entropy current and the form of the evolution equation $(s, \mathbf{j}_s, \mathcal{F})$. Therefore, the positive entropy production is supplemented by (22) in the Liu procedure.

$$\partial_t \boldsymbol{\xi} + \mathcal{F} = \mathbf{0},$$

$$\partial_1 s \partial_t \boldsymbol{\xi} + \partial_2 s \cdot \partial_t \nabla \boldsymbol{\xi} + \partial_3 s : \partial_t \nabla^2 \boldsymbol{\xi} + \partial_1 \mathbf{j}_s \cdot \nabla \boldsymbol{\xi} + \partial_2 \mathbf{j}_s : \nabla^2 \boldsymbol{\xi} + \partial_3 \mathbf{j}_s \cdot \nabla^3 \boldsymbol{\xi} \geq 0.$$

Here we applied the hypothesis of relocalizability, as in case of extended thermodynamic systems. According to Liu's theorem there exist a Γ , to be determined from the Liu equations, which can be written in a particularly simple form

$$\begin{aligned} \partial_1 s - \Gamma &= \mathbf{0}, \\ \partial_2 s &= \mathbf{0}, \\ \partial_3 s &= \mathbf{0}, \\ \partial_3 \mathbf{j}_s &= \mathbf{0}. \end{aligned}$$

The dissipation inequality in our case is

$$(23) \quad \partial_1 \mathbf{j}_s \cdot \nabla \boldsymbol{\xi} + \partial_2 \mathbf{j}_s : \nabla^2 \boldsymbol{\xi} - \Gamma \mathcal{F} \geq 0.$$

The solution of Liu equations gives that s depends only on the internal variable ξ , $\Gamma = Ds(\xi)$ is the derivative of the entropy and \mathbf{j}_s does not depend on the second gradient of ξ . Unfortunately these considerations do not simplify the entropy inequality at all, we should look for additional conditions. As previously we assume that ξ is a dynamic variables, Therefore there is no flow of entropy and the derivative of the entropy is zero if the internal variables are zero, that is $\mathbf{j}_s(\mathbf{0}, \mathbf{0}) = \mathbf{0}$ and $\Gamma(\mathbf{0}) = \mathbf{0}$. The first condition is quite natural. The second condition requires that the zero derivatives characterize the equilibrium of the material. Thus we can write the entropy current in the following form

$$\mathbf{j}_s(\xi, \nabla \xi) = \mathbf{A}(\xi, \nabla \xi) \Gamma(\xi).$$

This form of the entropy current is general with the assumptions above. As previously, we assume, that \mathbf{A} is a continuously differentiable function. In this case the entropy inequality turns out to have the form

$$(\nabla \cdot \mathbf{A} - \mathcal{F})\Gamma + \mathbf{A} \cdot \nabla \Gamma \geq 0.$$

Let us remark, that in (23) there are two constitutive quantities (the entropy current and \mathcal{F}) and three additive terms. To simplify the inequality for constructing a force current system there is no other choice that we have done here: we should unite two of the terms with some reasonable physical assumption. Now we can solve the resulted inequality and get an Onsagerian structure.

$$(24) \quad \nabla \cdot \mathbf{A} - \mathcal{F} = \mathbf{L}_{11}\Gamma + \mathbf{L}_{12}\nabla \Gamma,$$

$$(25) \quad \mathbf{A} = \mathbf{L}_{21}\Gamma + \mathbf{L}_{22}\nabla \Gamma.$$

Here \mathbf{L} is the positive semidefinite constitutive conductivity matrix. Eliminating \mathbf{A} from (24) and (25), we get

$$(26) \quad \partial_t \xi + \nabla \cdot (\mathbf{L}_{21}\Gamma + \mathbf{L}_{22}\nabla \Gamma) = \mathbf{L}_{11}\Gamma + \mathbf{L}_{12}\nabla \Gamma.$$

This is a balance equation with a special current and source term. To recognize the structure of the equation let us consider a single scalar internal variable ξ and isotropic material. In this case there is no cross effect between the thermodynamic forces of different tensorial order $L_{12} = 0$ and $\mathbf{L}_{21} = \mathbf{0}$, and the diagonal elements of the conductivity tensor \mathbf{L} are scalars $L_{11} = l_1$ and $\mathbf{L}_{22} = l_2 I$ (according to Curie principle or more properly the representation theorems of isotropic tensors). Therefore equation (26) simplifies to

$$(27) \quad \partial_t \xi = l_1 \Gamma - \nabla \cdot (l_2 \nabla \Gamma).$$

Here we have got an equation that is similar to the well known Ginzburg-Landau equation. However, there are some differences.

- In the second term of the right hand side under the space derivatives is Γ instead of ξ , contrary to the Ginzburg-Landau equation. The consequence of this difference is that the equation has the traditional homogeneous thermodynamic equilibrium solutions characterized by the condition $\Gamma = 0$.
- The sign of the material coefficients is determined by the Second Law, not by direct stability considerations.
- It is straightforward to extend the derivation to nonlinear and anisotropic cases. However, the nonlinearities and anisotropies show a different structure than in the original Ginzburg-Landau equation.

In the following (27) will be called *thermodynamic Ginzburg-Landau equation* [65]. This equation was derived by Verhás under some slightly different assumptions, as a governing equation for the transport of dynamic degrees of freedom [40, 66, 7].

Remark 2.1. *In all of the previous considerations the only mathematical step that seems to be restrictive from a physical point of view is the (continuous) differentiability of the current intensity factor. That condition can be weakened, but it will exclude the appearance of terms like \mathbf{j}_0 in case of classical and extended thermodynamics. The question is not mathematical and academic at all. We should not forget that the question of differentiability plays an important role in constitutive modelling and in thermodynamics in general. For example some phase boundaries are classified according to the differentiability of equilibrium state functions. Moreover, recently an experimental indication of phase boundaries was found also in the non-equilibrium state space, connected to internal variables. That concept makes possible the unification and the elaboration of a common thermodynamic frame for theories of failure and for theories of elasticity in mechanical continuum (e.g. in case of damaged brittle materials [67, 68]).*

2.4.2. Weakly nonlocal dynamic internal variable and extensive variable - Cahn-Hilliard equation. In our last example there is an extensive thermodynamic variable and also an internal one that is not the current density of the extensive as in extended irreversible thermodynamics. Therefore the basic state space of the physical quantities consists of the extensive quantity \mathbf{a} with a balance like evolution equation (2) and a dynamic internal variable $\boldsymbol{\xi}$ with an evolution equation (22), whose form is not specified. We are looking for nonlocal extension of the internal variable, therefore the constitutive space consists of the constitutive spaces of classical irreversible thermodynamics and the previous pure nonlocal internal variable theory together $C_{IE} = Z_a \times Z_\xi \times \text{Lin}(Z_a, \mathbb{R}^3) \times \text{Lin}(Z_\xi, \mathbb{R}^3) \times \text{Lin}(Z_\xi, \mathbb{R}^6)$. Our constitutive quantities are the entropy, the entropy current, the current of the extensives and the evolution equation of the internal variable $(s, \mathbf{j}_s, \mathbf{j}_a, \mathcal{F})$. All these quantities are functions defined on the constitutive space spanned by $(\mathbf{a}, \boldsymbol{\xi}, \nabla \mathbf{a}, \nabla \boldsymbol{\xi}, \nabla^2 \boldsymbol{\xi})$. We are looking for the restrictions on the constitutive quantities imposed by the non-negative entropy production (1) and the balance (2) with zero source term (pure dissipativity) and the condition of relocalizability. Considering the later we do not impose the space derivative of the evolution equation of the internal variables as an additional constraint. Because of the constitutive dependencies, the constraints and the entropy inequality can be written in the following form

$$\begin{aligned} \partial_t \mathbf{a} + \partial_1 \mathbf{j}_a \cdot \nabla \mathbf{a} + \partial_2 \mathbf{j}_a \cdot \nabla \boldsymbol{\xi} + \partial_3 \mathbf{j}_a : \nabla^2 \mathbf{a} + \partial_4 \mathbf{j}_a : \nabla^2 \boldsymbol{\xi} + \partial_5 \mathbf{j}_a \cdot \nabla^3 \boldsymbol{\xi} &= \mathbf{0}, \\ \partial_t \boldsymbol{\xi} + \mathcal{F} &= \mathbf{0}, \\ \partial_1 s \partial_t \mathbf{a} + \partial_2 s \partial_t \boldsymbol{\xi} + \partial_3 s \cdot \partial_t \nabla \mathbf{a} + \partial_4 s \cdot \partial_t \nabla \boldsymbol{\xi} + \partial_5 s : \partial_t \nabla^2 \boldsymbol{\xi} + \\ \partial_1 \mathbf{j}_s \cdot \nabla \mathbf{a} + \partial_2 \mathbf{j}_s \cdot \nabla \boldsymbol{\xi} + \partial_3 \mathbf{j}_s : \nabla^2 \mathbf{a} + \partial_4 \mathbf{j}_s : \nabla^2 \boldsymbol{\xi} + \partial_5 \mathbf{j}_s \cdot \nabla^3 \boldsymbol{\xi} &\geq 0. \end{aligned}$$

According to Liu's theorem there exist functions Γ_e and Γ_ξ to be determined from Liu equations written as

$$\begin{aligned} \partial_1 s - \Gamma_e &= \mathbf{0}, \\ \partial_2 s - \Gamma_\xi &= \mathbf{0}, \\ \partial_i s &= \mathbf{0}, \quad i = 3, 4, \\ (\partial_5 s)^s &= 0, \\ (\partial_3 \mathbf{j}_s - \Gamma_e \partial_3 \mathbf{j}_a)^s &= \mathbf{0}, \\ (\partial_5 \mathbf{j}_s - \Gamma_e \partial_5 \mathbf{j}_a)^s &= \mathbf{0}. \end{aligned}$$

Here the superscript s of the last equation can be expressed best with indexes that $(\partial_5)_{ijk}(\mathbf{j})_l$ is symmetric in ijk . A solution of the Liu equations results in that the entropy depends only on the extensive and on the internal variable. The derivative of the entropy gives the Lagrange multipliers $Ds(\mathbf{a}, \boldsymbol{\xi}) = (\partial_a s, \partial_\xi s) = (\Gamma_e, \Gamma_\xi)$. A consequence of the last two equations is

$$(28) \quad \mathbf{j}_s = \Gamma_e \mathbf{j}_a + \mathbf{j}_0(\mathbf{a}, \boldsymbol{\xi}, \nabla \boldsymbol{\xi}).$$

Here \mathbf{j}_0 is an arbitrary function of the variables denoted above. In the first term we can recognize the classical entropy current. This solution of the Liu equations simplifies the dissipation inequality to

$$(29) \quad \nabla \Gamma_e \cdot \mathbf{j}_a + \nabla \cdot \mathbf{j}_0 - \Gamma_\xi \mathcal{F} \geq 0.$$

This form is simple but unfortunately we cannot solve it without any further ado. We require that in case of zero internal variable the entropy flow reduces to the classical form. The dynamic property of the internal variable gives the additive term of the entropy current in the following form

$$\mathbf{j}_0(\mathbf{a}, \boldsymbol{\xi}, \nabla \boldsymbol{\xi}) = \mathbf{A}(\mathbf{a}, \boldsymbol{\xi}, \nabla \boldsymbol{\xi}) \Gamma_\xi.$$

Now the dissipation inequality transforms to

$$\mathbf{j}_a \cdot \nabla \Gamma_e + \mathbf{A} \cdot \nabla \Gamma_\xi + (\nabla \cdot \mathbf{A} - \mathcal{F}) \Gamma_\xi \geq 0.$$

As in the previous sections we reckon the entropy as a primary quantity. Therefore we arrived to a force-current system, to a sum of products of undetermined and given functions of the constitutive state. Therefore the general solution the inequality results in the Onsagerian conductivity equations in a quasilinear form, with a positive semidefinite conductivity matrix \mathbf{L} , depending on the constitutive state

$$(30) \quad \mathbf{j}_a = \mathbf{L}_{11} \nabla \Gamma + \mathbf{L}_{12} \nabla \Gamma_\xi + \mathbf{L}_{13} \Gamma_\xi,$$

$$(31) \quad \mathbf{A} = \mathbf{L}_{21} \nabla \Gamma + \mathbf{L}_{22} \nabla \Gamma_\xi + \mathbf{L}_{23} \Gamma_\xi,$$

$$(32) \quad \nabla \cdot \mathbf{A} - \mathcal{F} = \mathbf{L}_{11} \nabla \Gamma + \mathbf{L}_{12} \nabla \Gamma_\xi + \mathbf{L}_{13} \Gamma_\xi.$$

Substituting (30) into (2) and (31) into (32) we arrive the following system of transport like equations with a definite balance form

$$(33) \quad \partial_t \mathbf{a} + \nabla \cdot (\mathbf{L}_{11} \nabla \Gamma + \mathbf{L}_{12} \nabla \Gamma_\xi + \mathbf{L}_{13} \Gamma_\xi) = \mathbf{0},$$

$$(34) \quad \partial_t \boldsymbol{\xi} + \nabla \cdot (\mathbf{L}_{21} \nabla \Gamma + \mathbf{L}_{22} \nabla \Gamma_\xi + \mathbf{L}_{23} \Gamma_\xi) = \mathbf{L}_{11} \nabla \Gamma + \mathbf{L}_{12} \nabla \Gamma_\xi + \mathbf{L}_{13} \Gamma_\xi.$$

On the other hand this choice of the basic state space indicates that the above set of evolution equations can be considered as generalization of the Cahn-Hilliard equation. To see the connection one must eliminate the internal variable. Let

us consider a very special example, where our basic state space is spanned by a single scalar extensive a and a single scalar internal variable ξ . Furthermore, let be the material isotropic and all the material coefficients are constant (strictly linear approximation). Finally, let us write the entropy in the canonical form (14), but with constant inductivities m . Now there are two coupled terms in the linear laws, because the current of the extensives \mathbf{j}_a and the current multiplier \mathbf{A} have same tensorial order

$$(35) \quad \mathbf{j}_a = l_{11}\nabla\Gamma - l_{12}m\nabla\xi,$$

$$(36) \quad \mathbf{A} = l_{21}\nabla\Gamma - l_{22}m\nabla\xi,$$

$$(37) \quad \nabla \cdot \mathbf{A} - \mathcal{F} = -l_3m\xi.$$

Here the matrix of the conductivity coefficients is positive definite to ensure nonnegative entropy production ($l_3 \geq 0$, $l_{11} \geq 0$, $l_{22} \geq 0$ and $l_{11}l_{22} - l_{12}l_{21} \geq 0$). We can eliminate $\nabla\xi$ from the first two equations to get an expression of the current multiplier \mathbf{A} as:

$$\mathbf{A} = l_{22}l_{12}^{-1}\mathbf{j}_a + (l_{21} - l_{22}l_{12}^{-1}l_{11})\nabla\Gamma.$$

We can put this formula into (37) and so eliminating ξ from (35) we get

$$(38) \quad \mathbf{j}_a = l_{11}\nabla(\Gamma + \alpha\Delta\Gamma) + \beta\nabla\nabla \cdot \mathbf{j}_a + l_{12}l_3^{-1}\partial_t\xi,$$

where $\alpha = l_{11}^{-1}l_3^{-1}(l_{12}l_{21} - l_{22}l_{11})$ and $\beta = l_3^{-1}l_{22}$. we can neglect the last term in the above expression if $l_{12} = 0$ or the change of ξ is very slow. So we eliminate the internal variable and got (2) and (38) as a system of equations to be solved. After some straightforward manipulation we finally get

$$(39) \quad \partial_t a + l_{11}\Delta(\Gamma - \alpha\Delta\Gamma) - \beta\Delta\partial_t a = 0.$$

In our case, when a represents a single scalar physical quantity (e.g. mass density) then l_{11} , α and β are positive numbers. Equation (39) is similar to the Cahn-Hilliard equation, but not the same, therefore it will be called *thermodynamic Cahn-Hilliard equation*. It is worth to give a short comparison.

- The last extra term does not appear in the traditional equation. Gurtin derived a similar additional term with the help of microforce balance (a strong additional assumption) in the special case when the variable a is the mass density [64]. As regards some generalizations and the properties of the solution see [69, 70]. This term is similar that one could get in case of the Gurer-Krumhansl equation and there are several experimental indications for this kind of generalization with the Cahn-Hilliard equation, too [71]. Supposing that $l_{22} = 0$, this additional term vanishes. Let us recognize that this rate dependent term appears in the linear Onsagerian conductivity equations at the same thermodynamic approximation level as all the others.
- Instead of the extensive \mathbf{a} , the intensive variable Γ is under the second Laplace operator, contrary to the original Cahn-Hilliard equation. The equilibrium solutions of the thermodynamic Cahn-Hilliard equation are homogeneous.
- Every material parameter in (39) is positive, as a consequence of the Second Law, a nonnegative entropy production. In the original equation the signs of the coefficients are fixed according to stability considerations seemingly independently of the entropy balance.

- The nonlinearities and anisotropies show a different structure than in the original equation.
- In most of the previous derivations of the Cahn-Hilliard equation, a "generalized chemical potential" appeared by analogy, and was inserted without any further ado into the mass balance. Here in our derivation we have got a real generalization of the diffusion current in the same sense and with the same method as in the original irreversible thermodynamic approach.
- Treating the derivation of the thermodynamic Cahn-Hilliard equation a scalar extensive and a scalar internal variable was introduced. As \mathbf{a} can denote the Cartesian product of several extensives, the coupling in conductivity equations (35)-(37) can become more involved, even in the simplest isotropic case. On the other hand, the introduction of an internal variable with the tensorial order of the current (one order higher than the extensive) results in the very same structure of the final Cahn-Hilliard type equation.

3. HEURISTIC WEAKLY NONLOCAL IRREVERSIBLE THERMODYNAMICS

In the previous section weakly nonlocal thermodynamics was treated in a more or less mathematical level. Our point of view, the leading idea behind the previous section was to give the background and the conditions of applicability of the classical irreversible thermodynamic approach far beyond the validity of local equilibrium. We have seen, that the essence of the classical method of irreversible thermodynamics, the construction of proper force-current systems can be applied not only to systems with inertia, but for essentially nonlocal systems, too. Moreover, the local character of the theory can be preserved introducing special internal variables, current multipliers.

The general basic conditions to create solvable force-current systems were the following:

- (1) *Entropy* - there exist an extensive entropy with nonnegative production. Entropy and entropy current are constitutive functions. The entropy is the primary constitutive function of any thermodynamic theory.
- (2) *Pure dissipative systems* - the source term is zero in the balances of considered extensives.
- (3) *Relocalizability* - the entropy, as fundamental potential function expressing the properties of ordinary (equilibrium thermodynamic) systems does not depend on gradients of the basic variables.

In weakly nonlocal theories this is a postulate, in case of more restricted constitutive spaces is a consequence of the thermodynamic requirements as we have seen in the previous sections.

- (4) *Dynamic internal variables* - are those that vanish in (local) equilibrium. This assumption results in two consequences for sufficiently smooth constitutive functions:
 - If the entropy is a concave function of the internal variables (in the nonequilibrium part of the state space) then we arrive at the Gyarmati form of nonequilibrium entropy (14).
 - The Nyíri form generalized entropy current (18).

Here, in this section, based on the above properties, we treat some further weakly nonlocal relocalizable systems in a more heuristic way. This heuristic point of view shows, how one can get solvable (Liu) equations and solvable entropy inequality

in the more exact and formal mathematical procedure. In the first subsection we investigate the parade ground of thermodynamic theories and develop weakly non-local heat conduction. After that we show that a current multiplier of current multipliers can have an importance and derive Gurtin's term in Ginzburg-Landau equation. Finally we recall that current multipliers are not unique, there are several possibilities to transform the dissipation inequality into a solvable form. Here we investigate extended thermodynamics as a special internal variable theory and investigate the conditions to get a balance like evolution equation for the internal variables.

3.1. Weakly nonlocal heat conduction - Guyer-Krumhansl equation [72, 73]. The different special theories of thermodynamic origin as diffusion, mechanical, electromagnetic interactions, etc. all can show phenomena that can be explained by weakly nonlocal extensions of the classical equations. Let us investigate more closely the most frequently treated basic example of all thermodynamic investigations, the phenomena of heat conduction. Now the single extensive quantity is the internal energy, therefore $\mathbf{a} = (a_1) \equiv u$. Instead of citing the previously derived formulas we will apply a simplified, more heuristic procedure. The example demonstrates the role of the continuization hypotheses of local equilibrium, local state and relocalizability giving the different heat conduction theories respectively. We will see that the Guyer-Krumhansl equation is the thermodynamically consistent first weakly nonlocal extension of the Cattaneo-Vernotte equation and the structure of the equations is independent of the particular material properties.

The balance of internal energy is the starting point. Let us denote the heat current density by $\mathbf{j}_a = (\mathbf{j}_u \equiv) \mathbf{q}$ and we assume that the system is purely dissipative.

$$(40) \quad \partial_t u + \nabla \cdot \mathbf{q} = 0.$$

For the case of simplicity, we consider isotropic material and constant thermal inductivity \mathbf{m} . In this case $\mathbf{m} = mI$, therefore the entropy function (14) can be written as

$$(41) \quad s(u, \mathbf{q}) = s_0(u) - \frac{1}{2} m \mathbf{q}^2,$$

where $m > 0$. The Nyíri form of the entropy current is

$$(42) \quad \mathbf{j}_s = \mathbf{B}_u \cdot \mathbf{q}.$$

Now the entropy production (19) can be calculated by composite derivations

$$(43) \quad \begin{aligned} & \partial_t s + \nabla \cdot \mathbf{j}_s = \\ & \partial_u s \partial_t u + \partial_q s \partial_t \mathbf{q} + \nabla \cdot (\mathbf{B}_u \cdot \mathbf{q}) = \\ & (\mathbf{B}_u - \frac{1}{T} I) : \nabla \mathbf{q} + (\nabla \cdot \mathbf{B}_u - m \partial_t \mathbf{q}) \cdot \mathbf{q} \geq 0. \end{aligned}$$

We can recognize (19) in our special case. According to Curie principle (representation theorems of isotropic tensors, e.g. [74, 75]) there is no cross coupling between the different thermodynamic interactions. Moreover in (20) $\mathbf{L}_{11} = lI$, where l is a scalar and \mathbf{L}_{22} can be written with indices as

$$(L_{22})_{ijkl} = l_1 \delta_{ik} \delta_{jl} + l_2 \delta_{il} \delta_{jk} + l_3 \delta_{ij} \delta_{kl}.$$

Therefore the linear approximation of Onsager (20) reduces to

$$(44) \quad \begin{aligned} \mathbf{q} &= l(\nabla \cdot \mathbf{B}_u - m\partial_t \mathbf{q}) \\ \mathbf{B}_u - \frac{1}{T}\mathbf{I} &= l_1(\nabla \mathbf{q}) + l_2(\nabla \mathbf{q})^* + l_3 \nabla \cdot \mathbf{q} \mathbf{I}. \end{aligned}$$

where $*$ denotes the transpose. Eliminating \mathbf{B}_u we get

$$(45) \quad lm\partial_t \mathbf{q} + \mathbf{q} = l\nabla \frac{1}{T} + l(l_1 \Delta \mathbf{q} + (l_2 + l_3) \nabla^2 \cdot \mathbf{q}).$$

This is exactly the Guyer-Krumhansl equation for the heat current, introduced to describe the thermal properties of some crystals at low temperatures [72]. Originally it was derived using kinetic physics and later two-fluid hydrodynamics [76]. At the last decades there were several attempts to get the equation from pure non-equilibrium thermodynamics [77, 78, 79, 80, 81, 41, 82]. However, all of these derivations contain several ad-hoc assumptions, whose generalization for more difficult situations can be very ponderous if not impossible (e.g. two-fluid hydrodynamics is an internal variable theory from a thermodynamic point of view, but with a particular interpretation of the internal variables). One of these assumptions is a balance like dynamics for the heat current \mathbf{q} that is a result of the general structure in our case. If we do not eliminate \mathbf{B}_u from the material equations, then we can get a dynamic equation of the heat current that has a special balance form (more properly it will be an equation of Ginzburg-Landau type) indicating that the current intensity factor can be considered as a current density of the heat current \mathbf{q} [40]. We analyze further that property later.

In the special case of $l_1 = l_2 = l_3 = 0$, we get the Cattaneo-Vernotte equation for the heat current. The conductivity equations indicate clearly that the local state hypothesis is applied, the entropy current has its traditional form $\mathbf{j}_s = \mathbf{j}_q/T$, but the entropy itself depends on the dynamic nonequilibrium variable \mathbf{q} . The Cattaneo-Vernotte equation is a wave like equation for the heat conduction phenomena. It was Gyarmati who first derived it in the frame of irreversible thermodynamics with the help of entropy function (41) [47]. Further, we get the Fourier heat conduction equation when $m = 0$, in local equilibrium, when the entropy depends only on internal energy u .

In case of constant material coefficients (m, l, l_1, \dots) it is easy to derive a telegraph type equation directly for the temperature T . We should introduce the specific heat $u = cT$ and substitute (40) and its time derivative into the divergence of (45)

$$(46) \quad lmc\partial_{tt}T + c\partial_t T + l\Delta \frac{1}{T} + lc(l_1 + l_2 + l_3)\Delta \partial_t T = 0.$$

This reduction is remarkable and discussed from several points of view. First of all in several practical problems the material parameters cannot be considered as constant quantities (see e.g. [83]). On the other hand the reduction of a vector and a scalar equation (40) and (45) into a single scalar one results in a loss of information. The system (40) and (45) is equivalent to (46) if the heat current field is rotation free.

3.2. Nonlocal internal variables.

3.2.1. Generalized thermodynamic Ginzburg-Landau equation. In this subsection we develop the results of subsection 2.4.1 and show how can an additional rate term appear in the Ginzburg-Landau equation.

As previously our task is to find an evolution equation of an internal variable that corresponds to the requirement of nonnegative entropy production. The basic state space is the vector (moreover Banach) space of the variable $\boldsymbol{\xi}$ and we do not know a dynamic equation for the processes. We suppose a relocizable theory, therefore the entropy function depends only on the internal variable. We will use the notation $\Gamma_\xi := Ds(\boldsymbol{\xi})$. If $\boldsymbol{\xi} = \mathbf{0}$ then $\Gamma_\xi(\mathbf{0}) = \mathbf{0}$, because $\boldsymbol{\xi}$ is a dynamic variable. As regards the entropy current we apply the previous physical assumptions also in this case: if $\boldsymbol{\xi}$ was zero then there is no entropy flow. Moreover, in the light of this assumption and according to the mean value theorem, the entropy current can be written as a linear function of the derivative of the entropy, as in classical irreversible thermodynamics $\mathbf{j}_s = \mathbf{A}\Gamma_\xi$. Here we introduced a nonlocal internal variable, a current multiplier \mathbf{A} . We can go farther, and introduce the current multiplier into the basic state space. For the sake of simplicity, we suppose that the entropy function does not depend on \mathbf{A} , that is we are not interested in the associated memory effects, we are investigating only the nonlocal extension. The form of the entropy current is similar to that of the Cahn-Hilliard equation:

$$\mathbf{j}_s = \mathbf{A}\Gamma_\xi + \mathbf{B} \cdot \mathbf{A}.$$

Here \mathbf{B} is a current multiplier of the first current multiplier \mathbf{A} . This form expresses the physical assumption, that there is no entropy flow when the internal variables $\boldsymbol{\xi}$ and \mathbf{A} are zero. The tensorial order of \mathbf{A} is one order higher than the tensorial order of $\boldsymbol{\xi}$ and the tensorial order of \mathbf{B} is one order higher than the tensorial order of \mathbf{A} as it should be considering the most general linear connection. Therefore the entropy production follows as

$$(47) \quad \partial_t s(\boldsymbol{\xi}) + \nabla \cdot \mathbf{j}_s = (\partial_t \boldsymbol{\xi} + \nabla \cdot (\mathbf{A})\Gamma_\xi + \mathbf{A} \cdot (\nabla \Gamma_\xi + \nabla \cdot \mathbf{B}) + \mathbf{B} : \nabla \mathbf{A} \geq 0.$$

It is straightforward to put down the Onsagerian conductivity equations, but in general one cannot simplify them. Therefore, we will treat here only the simplest situation, when $\boldsymbol{\xi} = \xi$ is scalar, the material is isotropic and the approximation is strictly linear (the conductivity coefficients are constants). Now the conductivity equations are reduced to the following form

$$(48) \quad \partial_t \xi + \nabla \cdot \mathbf{A} = l_1 \Gamma_\xi$$

$$(49) \quad \mathbf{A} = l_2 (\nabla \Gamma_\xi + \nabla \cdot \mathbf{B})$$

$$(50) \quad \mathbf{B} = l_3^1 \nabla \mathbf{A} + l_3^2 (\nabla \mathbf{A})^* + l_3^3 \nabla \cdot \mathbf{A},$$

where $l_1, l_2, l_3^1, l_3^2, l_3^3$ are positive, scalar, constant coefficients. Now a simple calculation eliminates \mathbf{A} and \mathbf{B} from the above equations:

$$(51) \quad \partial_t \xi = l_1 \Gamma_\xi - l_2 \Delta (1 + l_1 l_3) \Gamma_\xi + l_3 \Delta \partial_t \xi.$$

Here $l_3 = l_3^1 + l_3^2 + l_3^3$. The last term, that is additional to (27) corresponds to the generalized Ginzburg-Landau equation of Gurtin. One can get back the thermodynamic Ginzburg-Landau equation (27) in the special case when $l_3^1 = l_3^2 = l_3^3 = 0$, that is $\mathbf{B} \equiv \mathbf{0}$. The positivity (positive definiteness in a more general situation) of the material coefficients is ensured by the Second Law. However, we should observe, that this generalization has not changed the characteristic thermodynamic term which differs from the original Ginzburg-Landau form: Γ_ξ stands under the Laplace operator instead of ξ .

We can continue the introduction of new nonlocal internal variables, putting \mathbf{B} into the basic state space. In this case \mathbf{B} becomes an internal variable and

we can introduce a corresponding current multiplier. Continuing this procedure, we can develop a whole phenomenological hierarchy of weakly nonlocal transport equations of higher and higher orders. The further research in this direction has a special importance for the kinetic theories. The outlined phenomenological hierarchy of nonlocal equations can suggest an approach similar to the momentum series expansion [84, 80]. On the other hand it is straightforward to extend the above treatment (with Liu procedure or without it) considering memory effects, too.

3.2.2. Are the dynamic equations of internal variables balances or not? In section 2.3 we have seen that in extended irreversible thermodynamics, when the currents of the extensives are the internal variables, the final constitutive-evolution equation of the currents did not have a balance form in general. Later, in section 2.4.1 for a pure internal variable the derived evolution equation (34) had a definite balance form. The reason of the difference is hidden in the way of the generalization of the entropy current. The classical form reads as a product of the currents of the extensives and the corresponding entropic intensives

$$(52) \quad \mathbf{j}_s = \partial_a s \mathbf{j}_a$$

In the generalization of Nyíri, in section 2.3 developing weakly nonlocal extended irreversible thermodynamics, the entropy current is assumed to be a homogeneous linear function of the currents of the extensives

$$(53) \quad \mathbf{j}_s = \mathbf{B} \cdot \mathbf{j}_a.$$

On the other hand, in case of dynamic variables that vanish in equilibrium and the entropy characterizes the equilibrium the entropy current was assumed to be a linear function of the entropic intensives, conjugated to the corresponding extensive variable.

$$(54) \quad \mathbf{j}_s = \mathbf{A} \cdot \partial_\xi s.$$

This form is a different generalization of (52) than (53) and was applied in section 2.3 related to model B, leading to the thermodynamic Cahn-Hilliard equation. Extended thermodynamics looks like a special case of the system treated in 2.3, because currents of extensives can be considered as special internal variables. How the two assumptions are related to each other? Could we arrive to an evolution equation with balance form also in case of extended thermodynamics as it is generally expected [39, 50]? To answer this question we should investigate the forms of the entropy inequality more closely. Instead of simply referring to the results of the previous sections we will give again a less refined, briefer, more direct, although sloppier, traditional irreversible thermodynamic approach.

To get the entropy production of weakly nonlocal extended thermodynamics (19) we consider a system with a basic state spanned by extensive \mathbf{a} and internal variable ξ together. In case of extended thermodynamics the (dynamic) internal variable is the (dynamic) current of the extensives $\xi = \mathbf{j}_a$. Assuming a local entropy function $s(\mathbf{a}, \mathbf{j}_a)$ one can get (19) by substituting (53), (10) and (2) into the entropy balance

as follows

$$\begin{aligned}
 \partial_t s + \nabla \cdot \mathbf{j}_s &= \\
 \partial_a s \cdot \partial_t \mathbf{a} + \partial_j s \cdot \partial_t \mathbf{j}_a + \nabla \cdot (\mathbf{B} \cdot \mathbf{j}_a) &= \\
 -\partial_a s \cdot \nabla \cdot \mathbf{j}_a - \partial_j s \cdot \mathcal{G} + \mathbf{B} : \nabla \mathbf{j}_a + \nabla \cdot \mathbf{B} \cdot \mathbf{j}_a &= \\
 (\mathbf{B} - \partial_a s \mathbf{I}) : \nabla \mathbf{j}_a + (\nabla \cdot \mathbf{B} + \mathcal{G} \hat{\mathbf{m}}) \cdot \mathbf{j}_a &\geq 0.
 \end{aligned}
 \tag{55}$$

Here we exploited the Gyarmati form entropy (14) and wrote $\partial_j s = -\hat{\mathbf{m}} \cdot \mathbf{j}_a$. Because the entropy function is the primary constitutive quantity we can conclude that (55) resulted in a solvable inequality for the two (!) constitutive functions \mathbf{B} and \mathcal{G} .

On the other hand, when the basic state space is the same as previously but with a general dynamic internal variable one evidently cannot assume any more that the entropy current is proportional to the dynamic variable and one cannot write the entropy current in a one term form of (54). However, a splitting into two terms, a classical and a non classical one, works well. Therefore we will assume the following configuration of the entropy current

$$\mathbf{j}_s = \partial_a s \mathbf{j}_a + \mathbf{B}' \cdot \boldsymbol{\xi}.
 \tag{56}$$

In this case we can go forward similarly to the previous derivation and substitute (2) and Gyarmati entropy into the balance of entropy to get

$$\begin{aligned}
 \partial_t s + \nabla \cdot \mathbf{j}_s &= \\
 \partial_a s \cdot \partial_t \mathbf{a} + \partial_\xi s \cdot \partial_t \boldsymbol{\xi} + \nabla \cdot (\partial_a s \mathbf{j}_a + \mathbf{B}' \cdot \boldsymbol{\xi}) &= \\
 -\partial_a s \cdot \nabla \cdot \mathbf{j}_a - \partial_\xi s \cdot \mathcal{F} + \partial_a s \nabla \cdot \mathbf{j}_a + \mathbf{j}_a \cdot \nabla \partial_a s + \mathbf{B}' : \nabla \boldsymbol{\xi} + \nabla \cdot \mathbf{B}' \cdot \boldsymbol{\xi} &= \\
 \mathbf{B}' : \nabla \boldsymbol{\xi} + \mathbf{j}_a \cdot \nabla \partial_a s + (\nabla \cdot \mathbf{B}' + \hat{\mathbf{m}} \mathcal{F}) \cdot \boldsymbol{\xi} &\geq 0.
 \end{aligned}
 \tag{57}$$

This is a solvable inequality for the three (!) constitutive functions \mathbf{B}' , \mathcal{F} and \mathbf{j}_a . Moreover it reduces to the previous (55) one in the special case of $\boldsymbol{\xi} = \mathbf{j}_a$, when $\mathcal{G} = \mathcal{F}$ and $\mathbf{B} = \mathbf{B}' + \partial_a s \mathbf{I}$. Therefore $\mathbf{j}_s = (\mathbf{B}' + \partial_a s \mathbf{I}) \cdot \mathbf{j}_a$. However, the solution of the inequality does not result in a balance form evolution equation for the internal variable, similarly to the extended thermodynamic case.

In section 2.4.2 the entropy current was assumed to have the following, different form

$$\mathbf{j}_s = \partial_a s \mathbf{j}_a + \mathbf{A} \cdot \partial_\xi s.
 \tag{58}$$

Now, we can substitute (2) and (28) into the balance of entropy to get the entropy inequality in the following configuration

$$\begin{aligned}
 \partial_t s + \nabla \cdot \mathbf{j}_s &= \\
 \partial_a s \cdot \partial_t \mathbf{a} + \partial_\xi s \cdot \partial_t \boldsymbol{\xi} + \nabla \cdot (\partial_a s \mathbf{j}_a + \mathbf{A} \cdot \partial_\xi s) &= \\
 -\partial_a s \cdot \nabla \cdot \mathbf{j}_a - \partial_\xi s \cdot \mathcal{F} + \partial_a s \nabla \cdot \mathbf{j}_a + \mathbf{j}_a \cdot \nabla \partial_a s + \mathbf{A} : \nabla \partial_\xi s + \nabla \cdot \mathbf{A} \cdot \partial_\xi s &= \\
 \mathbf{A} : \nabla \partial_\xi s + \mathbf{j}_a \cdot \nabla \partial_a s + (\nabla \cdot \mathbf{A} - \mathcal{F}) \cdot \partial_\xi s &\geq 0.
 \end{aligned}
 \tag{59}$$

This is a solvable inequality for the three (!) constitutive functions \mathbf{A} , \mathcal{F} and \mathbf{j}_a again. Moreover, as we have seen in section 2.4.2 the solution results in a balance form evolution equation of the internal variable without applying Gyarmati entropy.

In the special case of extended thermodynamics, when $\boldsymbol{\xi} = \mathbf{j}_a$ the entropy inequality transforms to

$$\sigma_s = \mathbf{A} : \nabla \partial_j s + \mathbf{j}_a \cdot \nabla \partial_a s + (\nabla \cdot \mathbf{A} - \mathcal{F}) \cdot \partial_j s \geq 0.$$

As one can see the above inequality does not have a general solution being a sum of three product terms with two constitutive functions. However, with Gyarmati entropy $\partial_j s = -\hat{\mathbf{m}} \cdot \mathbf{j}_a$ and the inequality further transforms to

$$\sigma_s = \hat{\mathbf{m}} \cdot \mathbf{A} \cdot \nabla \mathbf{j}_a + (\nabla \cdot (\partial_a s I - \hat{\mathbf{m}} \cdot \mathbf{A}) - \hat{\mathbf{m}} \cdot \mathcal{F}) \cdot \mathbf{j}_a \geq 0.$$

Here we can recognize the classical solvable extended thermodynamic form with $\mathbf{B}' = \hat{\mathbf{m}} \cdot \mathbf{A}$ and the entropy current in the Nyíri form as $\mathbf{j}_s = \mathbf{B} \cdot \mathbf{j}_a = (-\hat{\mathbf{m}} \cdot \mathbf{A} + \partial_a s I) \cdot \mathbf{j}_a$. The inequality was transformed into a solvable force-current system, but the balance form of the arising evolution equation of the internal variable is destroyed.

As a summary of the previous investigations we have seen that in case of dynamic internal variables in general the balance form of the evolution equation is a consequence of natural physical conditions but in case of extended thermodynamics the limited constitutive structure of the theory seem to destroy the balance form of the evolution equations as general solutions of the dissipation inequality.

Let us remark here, that the easier formal applicability and heuristic power of the classical irreversible thermodynamic method can support but cannot replace the deeper constitutive investigations of the first section. First of all because the postulates of the more exact method were clearer and weaker than the heuristic one (e.g. the form of the entropy current (28) was consequence of the Liu equations). On the other hand one should identify correctly the basic state space and the constitutive space to see beyond the rather formal treatment of current-force systems of irreversible thermodynamics.

4. SUMMARY, CONCLUSIONS AND DISCUSSION

In the previous sections we have seen that it is possible to extend irreversible thermodynamics to nonlocal phenomena with the help of generalized internal variables. The extension is based on the generalization of the entropy current. Partial differential equations were constructed constitutively as evolution equations of the internal variables by a force-current structure. The structure of nonlinearities, the sign of coefficients were determined by the requirement of nonnegative entropy production. We have investigated the thermodynamic background of several classical equations of mathematical physics containing higher order space derivatives.

In the first section we have seen that the choice of the variables, basic and constitutive state spaces are crucial steps in thermodynamic theory construction. Basic state spaces are spanned by the physical quantities for that one wants to find an evolution equation. Constitutive state spaces are spanned by variables of the basic state space and their derivatives, the ones, the constitutive functions are supposed to depend on. The extension of the constitutive state space determines whether an evolution equation of an extensive or an internal variable considers nonlocal (or memory) effects or not.

In this paper we investigated the nonlocal extension only of dynamic internal variables. According to this view we can call a basic internal state variable nonlocal if the constitutive space contains at least its second order space derivatives. The following table summarizes the hierarchy of continuum theories from that point of view. \mathbf{a} denotes the array of extensive variables and $\boldsymbol{\xi}$ an array of internal variables as above.

	Basic state	Constitutive state	
		Local	Nonlocal
Classical	\mathbf{a}	$(\mathbf{a}, \nabla \mathbf{a})$	
Extended	$(\mathbf{a}, \mathbf{j}_a)$	$(\mathbf{a}, \mathbf{j}_a, \nabla \mathbf{a})$	$(\mathbf{a}, \mathbf{j}_a, \nabla \mathbf{a}, \nabla \mathbf{j}_a, \nabla^2 \mathbf{j}_a)$
Internal variable	ξ	$(\xi, \nabla \xi)$	$(\xi, \nabla \xi, \nabla^2 \xi)$
Classical + internal variable	(\mathbf{a}, ξ)	$(\mathbf{a}, \xi, \nabla \mathbf{a})$	$(\mathbf{a}, \xi, \nabla \mathbf{a}, \nabla \xi, \nabla^2 \xi)$

- The first row of the table concerns classical irreversible thermodynamics where the basic state space is spanned by specific extensive quantities. Classical irreversible thermodynamics is a local theory, in the sense of local equilibrium. Investigating its nonlocal extensions one can easily conclude that in case of relocizable theories (when entropy depends only on the basic state), the structure of the equations does not change and only the classical constitutive functions become gradient dependent.
- In the second row, where the basic state space is spanned by the extensive variables \mathbf{a} and their (dynamic) current densities \mathbf{j}_a one get classical extended thermodynamics. Extending the constitutive space with the gradients of the current densities gives a nonlocal theory and leads to Guyer-Krumhansl-like equations.
- For pure weakly nonlocal dynamic internal variables ξ the nonlocal extension of the classical ordinary differential equations results in Ginzburg-Landau-like equations.
- An extensive variable with a weakly nonlocal dynamic internal variable leads to Cahn-Hilliard-like equations.

Our investigations were restricted to relocizable theories, where the nonlocality can be grasped through nonclassical internal variables, introduced into the entropy current. The examples can be continued, other kind of basic and constitutive states and couplings can lead to other weakly nonlocal thermodynamic compatible equations. However, we may not forget that there are genuine weakly nonlocal theories and equations, with large importance and ample experimental evidence. A typical example is the traditional, non thermodynamic Ginzburg-Landau equation. Fortunately our methods are applicable to embed also true weakly nonlocal equations into nonequilibrium thermodynamics. The key aspects of Liu's procedure regarding that question and the first investigations related to the compatibility of the Ginzburg-Landau equation with nonequilibrium thermodynamics are given in [85].

We can observe that the emerging classification scheme is more refined than was given by Penrose and Five [86], Hohenberg and Halperin [61] or Gurtin [64]. The investigation of several existing weakly nonlocal equations (e.g. different phase field equations, the complex Ginzburg-Landau or Kardar-Parisi-Zhang) from the point of view of nonequilibrium thermodynamics can reveal richer structures, new terms, clearer conditions, surprising interrelations, hence a broader range of applicability than usually considered.

In this paper it was shown that contrary to the common belief, irreversible thermodynamics can be extended far beyond the usual local equilibrium or local state

hypotheses, one can consider dissipation related to nonlocal effects, too. A rigorous treatment showed that the seemingly intuitive steps of irreversible thermodynamic modelling are well supported and explained with Liu procedure. Several interesting consequences, new terms in the traditional equations and possible sources of physical generalizations were explored. There are several experimental and theoretical evidences that the characteristic additional term, containing the time derivative of the Laplacian of the basic state variable ($\Delta\partial_t$), appears in all of the treated classical equations (Guyer-Krumhansl, Ginzburg-Landau and Cahn-Hilliard). That term cannot be derived from variational considerations without any further ado, because its Frechet (strong) derivative is not symmetric [87].

An other important remark, that the stability structure of nonequilibrium thermodynamics is preserved in the weakly nonlocal extension. Therefore the entropy can serve as an Liapunov functional with appropriate initial and boundary conditions. These possible boundary conditions are more general than that were given by Penrose and Fife in their investigations regarding the thermodynamic compatibility of Ginzburg-Landau and Cahn-Hilliard equations. The physical difference is that now Liapunov functions can be generated for several different open systems. This kind of generalization goes farther than it was possible in homogeneous systems (see e.g. [37]) and also requires a specification of the function spaces where the solutions of the differential equations are investigated.

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6. APPENDIX A: LIU-PROCEDURE

The Liu-procedure is based on an application of a theorem in linear algebra. That theorem can be considered as a consequence of the Farkas' lemma [88], well known in linear programming. More properly it is a special case of the so called affine Farkas' lemma [89] (that can be easily derived from the original Farkas' lemma [90]). The recognition of this connection between the Liu theorem and Farkas lemma is due to some recent researches of Kirchner and Hauser [91].

In this appendix we formulate and prove that theorem of Liu [22] in a simple and general form best suited for our purpose. The proof below is essentially that of Liu.

\mathbb{V} and \mathbb{V}' are finite dimensional vector spaces, \mathbb{V}^* and \mathbb{V}'^* denote their duals.

Theorem 6.1. (*Liu*) $\mathbf{A} \in \text{Lin}(\mathbb{V}, \mathbb{V}')$, $\mathbf{B} \in \mathbb{V}'$, $\mathbf{a} \in \mathbb{V}^*$ and $b \in \mathbb{R}$. Then $\mathbf{a} \cdot \mathbf{x} + b \geq 0$ holds for all $\mathbf{x} \in \mathbb{V}$ such that $\mathbf{A} \cdot \mathbf{x} + \mathbf{B} = \mathbf{0}_{\mathbb{V}'}$ if and only if there is a $\lambda \in \mathbb{V}'^*$ such that

$$(60) \quad \mathbf{a} - \lambda \cdot \mathbf{A} = \mathbf{0}_{\mathbb{V}^*},$$

$$(61) \quad b - \lambda \cdot \mathbf{B} \geq 0.$$

(60) is called Liu-equation and (61) is the dissipation inequality in the thermodynamic literature.

Proof:

The backward direction is easier. If there is a λ with the above property, then for all $\mathbf{x} \in \mathbb{V}$, $0 \leq (\mathbf{a} - \lambda \cdot \mathbf{A}) \cdot \mathbf{x} - \lambda \cdot \mathbf{B} + b = (\mathbf{a} \cdot \mathbf{x} + b) - \lambda \cdot (\mathbf{A} \cdot \mathbf{x} + \mathbf{B})$ and the statement follows.

Now let us prove the forward direction. We introduce the notation $\mathcal{Aff} = \{\mathbf{x} \in \mathbb{V} \mid \mathbf{A} \cdot \mathbf{x} + \mathbf{B} = \mathbf{0}_{\mathbb{V}'}\}$. First of all we should see that $\text{Ker} \mathbf{A} \subset \text{Ker} \mathbf{a}$. Really if $\mathbf{y} \in \text{Ker} \mathbf{A}$ and $\mathbf{x} \in \mathcal{Aff}$ then for all $r \in \mathbb{R}$, $\mathbf{x} + r\mathbf{y} \in \mathcal{Aff}$, therefore according to the conditions of the theorem $0 \leq \mathbf{a} \cdot (\mathbf{x} + r\mathbf{y}) + b = \mathbf{a} \cdot \mathbf{x} + b + r\mathbf{a} \cdot \mathbf{y}$. For arbitrary r the inequality is true if and only if $\mathbf{a} \cdot \mathbf{y} = 0$ therefore $\text{Ker} \mathbf{A} \subset \text{Ker} \mathbf{a}$.

For the annullators of the kernels

$$(\text{Ker} \mathbf{a})^\perp \subset (\text{Ker} \mathbf{A})^\perp,$$

because $\forall \mathbf{x} \in \text{Ker} \mathbf{A} \subset \text{Ker} \mathbf{a}$, therefore, if $\mathbf{p} \in (\text{Ker} \mathbf{a})^\perp$ then $\mathbf{p} \cdot \mathbf{x} = 0$.

On the other hand $\text{Ker} \mathbf{A}^* = (\text{Ran} \mathbf{A})^\perp$, because if $\forall \mathbf{x} \in \mathbb{V} \quad \mathbf{y} \cdot \mathbf{A} \cdot \mathbf{x} = 0 = \mathbf{x} \cdot \mathbf{A}^* \cdot \mathbf{y}$ then from the left hand side of the equality $\mathbf{y} \in (\text{Ran} \mathbf{A})^\perp$ and from the right hand side $\mathbf{y} \in \text{Ker} \mathbf{A}^*$. For finite dimensional \mathbb{V} , $\mathbf{A}^{**} = \mathbf{A}$ and $(\mathbf{A}^\perp)^\perp = \mathbf{A}$, therefore we can transform the equality into the form

$$(\text{Ker} \mathbf{A})^\perp = \text{Ran} \mathbf{A}^*.$$

Putting together all these statements we can see, that $\mathbf{a} \in (\text{Ker} \mathbf{a})^\perp \subset (\text{Ker} \mathbf{A})^\perp = \text{Ran} \mathbf{A}^*$, therefore there is a $\lambda \in \mathbb{V}'^*$ such that $\mathbf{a} = \mathbf{A}^* \cdot \lambda$. In finite dimension follows (60). Therefore we can write for all \mathbf{x} with $\mathbf{A} \cdot \mathbf{x} + \mathbf{B} = \mathbf{0}_{\mathbb{V}'}$ that $0 \leq \mathbf{a} \cdot \mathbf{x} + b = \lambda \cdot \mathbf{A} \cdot \mathbf{x} + b = b - \lambda \cdot \mathbf{B}$.

The proof is complete.

Remark 6.1. If \mathbf{A} is surjective ($\dim(\text{Ran} \mathbf{A}) = \dim \mathbb{V}'$) then λ is unique.

7. APPENDIX B: MEAN-VALUE THEOREM

We applied Lagrange's mean value theorem several times in the paper. Here we formulate it in a way that is sufficiently general for our purpose. One can find a proof for example in [92].

Theorem 7.1. (Lagrange) Let \mathbb{X} be a normed space. $F : \mathbb{X} \rightarrow \mathbb{R}$, $\mathbf{x}, \mathbf{y} \in \mathbb{X}$. If F is differentiable on the closed section $[\mathbf{x}, \mathbf{y}]$, then there is a $\mathbf{z} \in [\mathbf{x}, \mathbf{y}]$, so that

$$F(\mathbf{y}) - F(\mathbf{x}) = DF(\mathbf{z}) \cdot (\mathbf{y} - \mathbf{x}),$$

where $DF(\mathbf{z})$ denotes the derivative of F at \mathbf{z} .

8. APPENDIX C: SOLUTION OF THE DISSIPATION INEQUALITY

In a force-current form the dissipation inequality can be written as a sum of the product of constitutive quantities and variables in the constitutive space. In this case a solution can be given in the form of the (nonlinear) Onsagerian conductivity relations. we formulate the statement and give the simple proof following Gurtin [64].

Theorem 8.1. Let \mathbb{V} be a finite dimensional vector space. $\mathbf{J} : \mathbb{V} \rightarrow \mathbb{V}^*$, $\mathbf{x} \mapsto \mathbf{J}(\mathbf{x})$ is a continuously differentiable function. If $\mathbf{J}(\mathbf{x}) \cdot \mathbf{x} \geq 0$ for all $\mathbf{x} \in \mathbb{V}$ then $\mathbf{J}(\mathbf{x}) = \mathbf{L}(\mathbf{x})\mathbf{x}$ where $\mathbf{L} : \mathbb{V} \rightarrow \text{Bilin}(\mathbb{V})$.

Proof:

From the inequality follows that for all positive number λ and $\mathbf{x} \in \mathbb{V}$, $\mathbf{J}(\lambda\mathbf{x}) \cdot \lambda\mathbf{x} \geq 0$, therefore $\mathbf{J}(\lambda\mathbf{x}) \cdot \mathbf{x} \geq 0$. In the limit when λ goes to zero we get that $\mathbf{J}(\mathbf{0}) \cdot \mathbf{x} \geq 0$.

That can be true for all $\mathbf{x} \in U$ if and only if $\mathbf{J}(\mathbf{0}) = \mathbf{0}$. The statement follows according to the mean value theorem.

Remark 8.1. *The structure is not unique in case of nonlinear relations.*

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